We sincerely thank both reviewers for thoroughly reading and commenting on our manuscript. Our answers, in normal text, are presented below the comments, which are bold. Changes to the manuscript are in italic with line numbers of the modified manuscript.

REVIEWER #1

The work presented by Ahlberg and coauthors investigates the variability of black carbon properties at an urban and rural site in southern Sweden. This manuscript fulfils the requirements of a "Measurement report" since it treats specific aerosol measurements and processes confined in a restricted area and time. However, the manuscript is structured as a full "Research Article" and falls a bit short on certain aspects of data analysis and interpretation. The final consequence is that the motivations, methods and goals are not always clear. I recommend major revision and resubmission. I hope that the major and specific comments listed below will help the authors in the rebuttal process.

Regarding the manuscript type, it is our understanding that the structure of a measurement report is very similar to a research article. If certain sections should be modified we hope to get feedback from the editor. With the answers below we hope that the methods and goals have been clarified.

MAJOR COMMENTS

First, there is an evident problem with sections and subsections. The article is organized without subsections; thus the assimilation of the scientific message becomes particularly complicated. The structure should be modified including subsections in the "methods" and result "sections".

We agree and have included subsections in section 2.2 (Instrumentation) and in the results in the modified manuscript to help the readers.

The overall motivation behind the paper is unclear. The authors mentioned climatic and health implications, but these are described very generally without a local (Swedish) perspective. The manuscript does not provide enough measurement time to address climatic issues but could draw a nice, even if very short, picture of air quality. I think this should be the redline of the entire manuscript and should include as motivation: the health impact of aerosol emission in Sweden (e.g. death per year), the history of Swedish reduction strategies and subsequent effects.

Although all field campaigns are merely snapshots of current air quality, we believe the reported results on BC size and number are the first of its kind in the region, and are therefore important to report for their influence on climate. Further, the importance of long-range transport for Swedish background concentrations are shown using a longer dataset. An epidemiological study on BC emissions in Sweden vs long range transport would be interesting but is a totally different study that could build on our results. To make the overall motivation behind the study clearer, we have changed the last paragraph of the introduction:

L52. The aim of this study was to compare BC particle properties in nearby urban and rural settings, and to investigate the influence of urban emissions on the rural background air. BC was measured using single particle laser induced incandescence and filter-based absorption. We compare BC mass and number concentrations, size distributions and mixing state. Furthermore, the relation to total particle number concentrations and chemically resolved PM1 are assessed. Trajectory analysis was used to assess the influence of long- and short-range transport.

There is nothing that can be practically done about this, but the fact that the urban and background measurements are not simultaneous is the weakest point of the manuscript. The authors should convince the reader that the background aerosol population do not change drastically from July to October. Till that point, the results shown in Figures 1,2,3 could be affected by many atmospheric processes such as precipitation, and changes in emission large scale circulation. Potentially due to this problem, the goal of the authors is not always clear.

We agree that it is unfortunate that all measurements could not be performed simultaneously since only one SP2 instrument was available. But to us, Figure 1 and 3 clearly shows that the diel cycle of BC properties in the urban environment is due to the local emissions. To compare the two periods further, we added the following

figure to the supplement, showing observed and modelled eBC at the rural background station during both the rural and urban campaign. Sources were modelled using ADCHEM (described in the manuscript) and standard CAMS emissions.

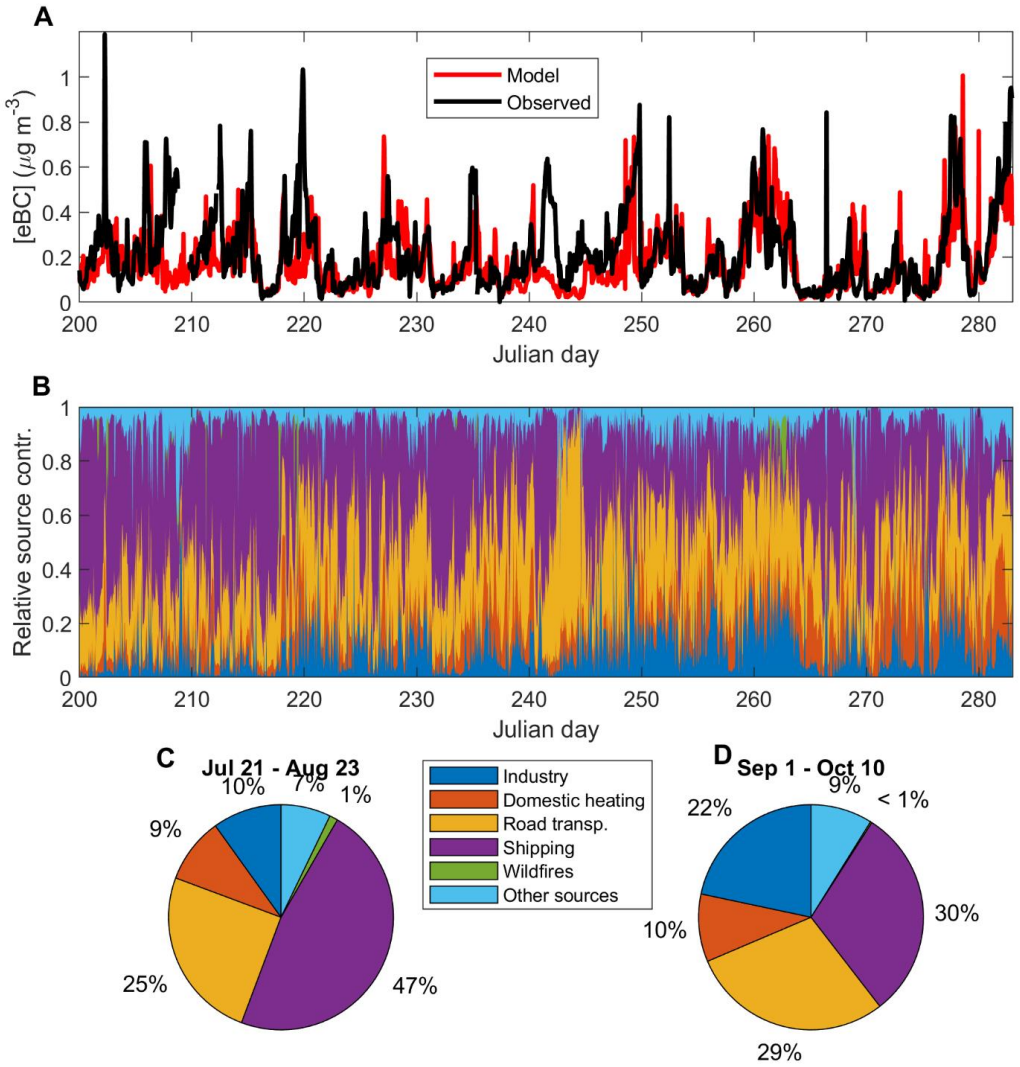


Figure S1. Modelled BC and measured eBC concentrations at the urban site during both campaigns (panel A), and modelled sources (panel B) using CAMS emission data and ADCHEM. Pie charts show the sources at the rural station during the respective campaigns.

The figure shows that the eBC concentrations in the background air did not vary much between the two campaigns, although the sources are slightly different. Hence, the background size distribution could also have changed. However, to us, the diel variation of GMD in figure 3 convincingly shows that the BC core size is driven by local traffic emissions and not seasonal change. The nighttime rBC GMD during the urban campaign is very similar to the GMD duringthe rural campaign.

Changes to the manuscript:

L62. Figure S1 shows that the BC concentrations at the rural site during July-October did not change drastically between the times of the two campaigns.

L193. The long-distance transported BC source contributions during the rural and urban measurement campaigns were estimated using the Lagrangian 1D-column chemistry transport model ADCHEM (Roldin et al., 2011; Roldin et al., 2019). ADCHEM was setup and run forward in time along pre-computed 14-days backward HYSPLIT air mass trajectories arriving 100 m a.g.l. at Hyltemossa, one new trajectory every hour. Source specific anthropogenic BC emissions along the trajectories were taken from the CAMS-GLOB-ANT v4.2 global emission inventory, which has a spatial resolution of 0.1°x0.1° (Granier et al., 2019). BC emissions from wildfires were estimated using the GFED4 emission inventory (Randerson et al., 2018).

Granier, C., Darras, S., Denier van der Gon, H., Doubalova, J., Elguindi, N., Galle, B., Gauss, M., Guevara, M., Jalkanen, J.-P., Kuenen, J., Liousse, C., Quack, B., Simpson, D., Sindelarova, K., The Copernicus Atmosphere Monitoring Service global and regional emissions (April 2019 version), Copernicus Atmosphere Monitoring Service (CAMS) report, doi:10.24380/d0bn-kx16, 2019.

Roldin, P., Swietlicki, E., Schurgers, G., Arneth, A., Lehtinen, K. E. J., Boy, M., and Kulmala, M.: Development and evaluation of the aerosol dynamics and gas phase chemistry model ADCHEM, Atmos. Chem. Phys., 11, 5867–5896, https://doi.org/10.5194/acp-11-5867-2011, 2011.

Roldin, P., Ehn, M., Kurtén, T., Olenius, T., Rissanen, M. P., Sarnela, N., Elm, J., Rantala, P., Hao, L., Hyttinen, N., Heikkinen, L., Worsnop, D. R., Pichelstorfer, L., Xavier, C., Clusius, P., Öström, E., Petäjä, T., Kulmala, M., Vehkamäki, H., Virtanen, A., Riipinen, I., and Boy, M.: The role of highly oxygenated organic molecules in the Boreal aerosol-cloud-climate system, Nat. Commun., 10, 4370, https://doi.org/10.1038/s41467-019-12338-8, 2019.

Randerson, J.T., van der Werf, G.R., Giglio, L., Collatz, G.J., and Kasibhatl, P.S., Global Fire Emissions Database, Version 4.1 (GFEDv4). ORNL DAAC, Oak Ridge, Tennessee, USA. https://doi.org/10.3334/ORNLDAAC/1293, 2018.

L258. Figure S1 shows that while BC concentrations at the rural site were similar during the two campaigns, the source sectors changed slightly. Notably, more industrial emissions and less shipping emissions affected the concentrations during the urban campaign.

Technically speaking there are some additional soft spots. It is very much not clear how the absorption data were treated and corrected, as a consequence the data are quite questionable. See specific comments. Moreover, the Aethalometer data are not essential to the scope of the paper since the SP2 provides mass concentration, size distribution and mixing state proxy.

See answers for specific comments below to the technical questions. The aethalometer data is essential to strengthen our view on the importance of long range transport since we used a much longer dataset. Also, we think that the discrepancy between rBC and eBC measurements is of interest to the community, and should be investigated further.

It must be described more clearly when ACTRIS data (absorption and chemical composition) were used. The reader realizes only towards the end of the manuscript that a year-long dataset was used. No description or introduction to these data is ever given.

We have clarified what data was used with the following graphics and comments:

L77. Figure 1 shows during which time the urban and rural campaigns took place and which data was used. BC properties and concentrations was determined using a Single Particle Soot Photometer (SP2) that was moved between the sites and Aethalometers (AE33) deployed permanently. Aethalometer data for the whole year was used from the rural site to assess source regions.

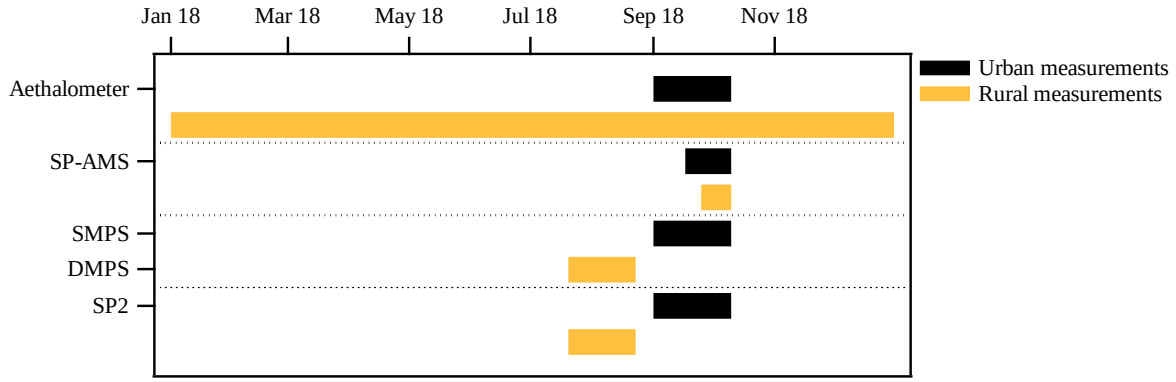


Figure 1. Overview of the data and instruments used.

SPECIFIC COMMENTS

L17: health effect is mentioned also in the introduction, but it is never really explained.

This is beyond the scope of this paper, and we refer the readers to the cited literature.

L34 It is an odd way to start a paper. I would remove the first sentence since it might apply to all atmospheric species.

Although a platitude to aerosol scientists, it is our way of saying that soot is ubiquitous, which is a common way to introduce and motivate research on BC.

L36: this is the motivation of your work and also the first sentence of the abstract. Still, I have zero ideas about how BC affects the climate and human health.

The statements have references to back it up. We have added WHO global air quality guidelines 2021 as a reference to further strengthen the claims with recent findings.

L40-41: I would provide an example for all properties or not report any example. Listing only the lensing effect does not add any relevant information since absorption enhancement is not a topic of the paper.

Agree. We removed the sentence.

L46-47: it appears like the reference for transport and deposited dose is missing.

The sentence now reads (with an added reference for experimental deposition of diesel exhaust particles):

L45. BC from different sources have different size distributions (Schwarz et al., 2008; Laborde et al., 2013; Saarikoski et al., 2021), that affects both transport (lifetime) and light interactions (Hinds, 2012), as well as deposited dose (Alfoldy et al., 2009; Rissler et al. 2012).

Rissler, J., E. Swietlicki, A. Bengtsson, C. Boman, J. Pagels, T. Sandström, A. Blomberg and J. Löndahl: Experimental determination of deposition of diesel exhaust particles in the human respiratory tract, Journal of Aerosol Science, 48, 18-33, 2012.

L50: Health and toxicity are mentioned a couple of times, but it is not yet explained how fundamental properties are connected to health.

We refer the interested readers to plenty of well cited literature e.g. WHO, 2021; WHO, 2013; IARC, 2014.

L55-56: main take-home message is summarized here. I think it does not belong to the introduction. Potentially makes more sense as a final statement of the abstract.

We agree and moved this sentence to the abstract.

L60-61: Please provide some evidence supporting your statement. July September is a long-time gap. I expect different dilution due to boundary layer height (I imagine lower temperature in October), different precipitation and washout, or different chemistry due to shorter sunlight duration…

See answer and graphic to major comment regarding figures 1,2 and 3 above.

L71-72: SP2 is not yet introduced. Move this last sentence to the instrumentation section. Since you mention turbulent flux. Can you estimate a loss fraction?

We want to keep the description of inlets in this section so we changed the sentence to:

L72. The single particle soot measurements at the rural site were conducted through a ¼'' tube at 20 liters per minute (turbulent flow), due to simultaneous Eddy Covariance flux measurements (not described here).

The loss estimations are described in Section 3.2 and shown in the supplement.

L74: This chapter is extremely long and complex. I would add a table listing the deployed instruments and measured variables at the two sites.

We agree and have added the graphic above showing the different instruments and when they were used as well as subsections for the various instruments.

L80: is good practice to provide the name, city and country of the manufacturer. Missing everywhere in the manuscript.

We do not believe ACP has this as a house standard rule. We prefer without.

L87: is this the saving rate of the SP2? A person not familiar with the SP2 would not understand what was does it mean. I think is not so important to be mentioned.

Typically saving raw data signals from all particles gives too much data to handle by the analysis program. This does not affect the concentration estimations but is important for at what timescale it is feasible to extract a size distribution. We'd like to keep this description so that SP2 users knows the settings used.

L90-91: What do you mean by "not satisfactory"? Was the reason connected to a wrong sizing of the DMA or a decrease in the performances of the SP2 (decreasing laser power, misalignment)? Are then the data valid?

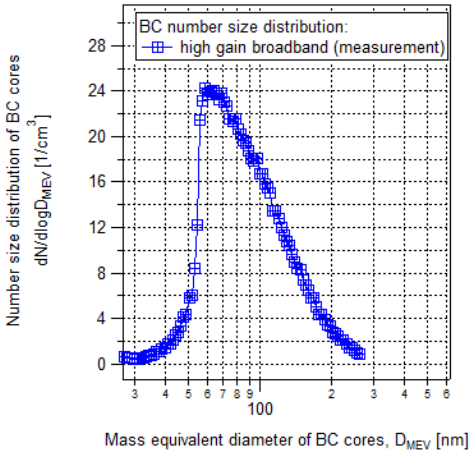
The calibration data of the rural campaign is missing due to a savings setting (human error). The instrument was calibrated before and after the campaign with similar results. Although this is definitely not ideal we strongly believe that the rBC data is valid. The instrument settings and alignments prior to each campaign ensures similar LII signals for a given mass. For instance, if the laser deteriorates for any reason, the current is typically amplified so that the power read by the output coupler stays constant. We changed a sentence to clarify:

L100. The response of the broadband incandescence detector from typical ambient BC particle masses varied with less than ±15 % for calibrations performed before and after the rural campaign.

L98: 62 nm is a very small diameter, but those particles will not contribute significantly to the total mass. My issue with this choice is that most, if not the totality, of previous SP2 paper, reports rBC particles

starting from 80-90 nm…which I might consider the safe side. Considering that you do not provide any counting efficiency, I do not understand why you chose such a low cut-off.

The cut off was chosen during analysis by setting the level of quantification threshold of the incandescence channel to the point where there's a sharp decrease in concentration. See figure below for an example. Since we don't make use of the number size distribution modes it doesn't matter for those results. For the number concentrations and fractions we discuss the limitations of the instrument and include the particles that we are certain are real. From personal experience with our SP2 as well as from intercomparison with other groups a D50 between 60-70 nm in mass equivalent diameter is quite normal.



L104: the coating is not directly measured by these two detectors. I would use caution with these statements since people might think that the SP2 directly provides coating thickness of BC-containing particles, which is well far from reality.

Agree. Changed to:

L112. BC particle coatings can be estimated by the SP2 using a separate scattering detector.

L106-111: I am genuinely confused by this explanation. Are talking of delay-time or LEO-fit. To me, it appears like a mixture of the two. Please rewrite it. If the position-sensitive detector was not used is not worth mentioning, since it adds confusion.

It is not easy to explain this without going into a great deal of detail, that is outside the scope of the manuscript. Instead we refer the reader to the cited literature. But we do say that we use the delay-time method and there is no mention of LEO or split-detector in the manuscript, so we don't know why there is confusion.

L114: since you mention Weingartner…what Cref value was used? Was it calculated for this specific aethalometer or taken from Weingartner or Collaud-Coen or Zanatta? These papers are based on AE31 though and not AE33.

Cref was not changed from the default value of 1.39 for the filters used. We added the following parenthesis:

L123. Default corrections for filter scattering (Cref = 1.39) and loading effects were used (Weingartner et al., 2003).

L125-133: Please add the chemical species identified by the SP-AMS. If all presented rBC mass is derived from the SP2 the calibration for refractory material is even described? In what sense the SP-AMS at the urban site did not provide robust results (instrument malfunction, wrong calibration)? Is this the reason why all AMS graphs at the urban site are plotted with arbitrary units? I have the feeling that this issue and the SP2 calibration problem (L90) undermine the credibility (accuracy, reproducibility) of the dataset and, as a consequence, the full manuscript. The authors should explain in more detail why and how the SP2 and SP-AMS data are still reliable despite the technical issues.

We added the species in the following sentence:

L133. The (non-refractory) components of particles in the air beam are vaporized on a heated tungsten plate (600 °C), subjected to 70 eV electron ionization, and the ions produced are detected and categorized (organic, nitrate, sulfate, ammonium or chloride) by a high-resolution time-of-flight mass spectrometer.

We also removed the description of rBC calibration of the SP-AMS.

For the SP-AMS, there was confusion around the measured mass loadings (based on field calibration with 300 nm mobility diameter ammonium nitrate particles). The rural loadings were higher throughout the campaign, despite very similar time trends for all non-refractory species (see Figure 7). We found the concentration in the rural data well supported by DMPS (same site) and FIDAS (nearby background site). We do not have measurements for a similar evaluation of the urban SPAMS. Hence, the combined evidence suggests the urban absolute concentrations from SPAMS are too low, which we attribute to calibration issues. Notably this would not have been detected if we were not comparing with another SPAMS downwind (at the rural site) which was properly supported by auxiliary measurements.

That the SP2 data is valid we hope is convincing by the answer to comment on L90-91 above. For the SP-AMS data, we simply use the fractions of chemical species and not the absolute mass concentrations in our interpretations.

L145: just a comment to point out that 64 nm of electric-mobility diameter does not correspond to 64 nm of mass equivalent diameter.

True. We removed that part in the methods section. We choose this value as a conservative minimum. It is tricky to compare the two instruments without a common size selection. In the results we also compare the number fractions using a larger diameter, to give the reader a range.

L171: back trajectories are not shown

Showing an example trajectory would not add to the manuscript.

RESULTS AND DISCUSSION

L183-191: There is no context to your observation. Up to me, these are low concentration for being an urban site.

It is true that the concentrations in Malmö are not that high compared to many other cities around the world. However, comparing Malmö to other sites and a wider context using only this short-term campaign would not suffice. We want to compare the nearby urban and rural sites only.

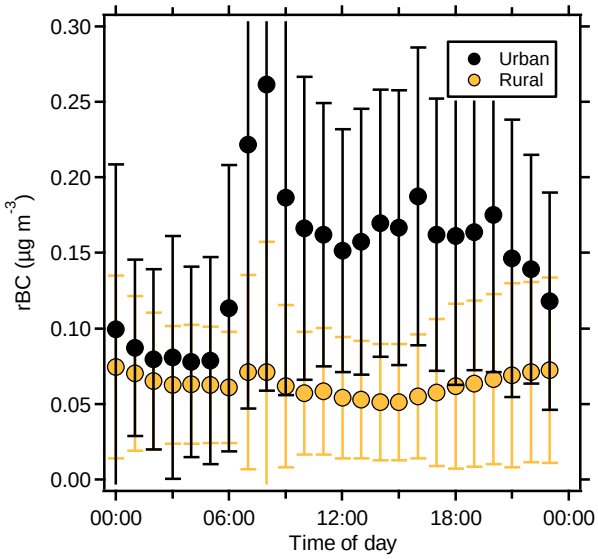
L186: it is already clear from its concentration that the curbside is not extremely polluted. At what percentage difference you would define extreme pollution? And based on what process?

True, we compared the two sites to show that the curbside was representative of the city. We removed the word exreme.

L209. This suggests that the curbside measurements are indicative for the city.

F1: provide error bars. Does the analysis include the weekends?

We added error bars, see new figure below. Yes, weekends are included.



L191: MAC of BC is not the only reason for the difference in AAE. Different AAE might be caused by a change in the chemical composition of absorbing aerosol or a change in the relative concentration of aerosol absorbing more light at the lower wavelength. If this is the case, MACbc will remain the same while AAE of total aerosol will increase.

True. We rephrased the sentence:

L214. The AAE is similar between the sites, with small differences likely owing to different BC sources.

L200-212: As it is also pointed out in the text, the rBC number fraction mostly depends on the diameter quantification limits of both the SP2 and especially DMA rather than on aerosol properties. So, I am not sure what should I retain out of this subchapter.

We agree that these are not hard numbers, but still think it is important. The lower limit is definitely too low since fresh BC particles without coating would have a larger mobility diameter than its mass equivalent diameter and the SP2 may not have 100% counting efficiency. The larger limit assumes that all BC particles counted by the SP2 are at least 130 nm in mobility diameter which is likely closer to the truth given the structure of fresh soot agglomerates. However, collapsed particles with little coating might be unaccounted for by this higher limit. The limits we choose give an estimate of BC number fractions between 2.7-6 % and 13.4-45 % for the rural and urban sites respectively. We think this difference is interesting and something that can be compared to in future studies.

L213-223: Here several factors must be considered and I strongly believe that the SP2 size cut is not the problem. Even if the SP2 size range could be easily accounted for by fitting a lognormal to the size distribution. Anyhow, no details are reported about the correction used for the AE-33 and I believe the problems are more connected with absorption calculation rather than the SP2 detection range. I recalculated the MAC from the values reported in Table 1. I recalculated Babs by multiplying Mebc by 7.77 m2/g. Then I calculated MAC as the ratio of Babs and MrBC. So, I obtain MAC values above 25 m2/g. This value is very similar to the mass attenuation coefficient used in the past to convert directly attenuation coefficients to eBC in the old AE31. So, I cannot say what happened here exactly, but I think that no Cref or correction was applied. I think you need to do a bit of extra thinking here and reconsider the relevance and accuracy of eBC measurements.

We used the default AE33 settings that is commonly used to report BC values and recommended by e.g. ACTRIS (https://www.actris-ecac.eu/particle-light-absorption.html). Since we report the MAC that was used anyone, as you did, can recalculate the Babs values. We do not want to use the SP2 rBC as a reference to report some equivalent rBC from the Aethalometer. As pointed out above, the correct Cref value was used. The difference between different BC instruments is ongoing research, e.g. ACTRIS now recommends a "Harmonization factor" to adjust AE33 Babs values. This a very important and interesting discussion and as the references in this paragraph show, we are not the first to report this difference.

L2017: was the 10-20% calculated or is it just a gentle guess? If it is calculated why is not applied to the measurements? Lensing effect cannot be excluded, but I hardly think that this is the main reason behind the eBC-rBC difference: You are using a MAC 1.6 times smaller than Swedish ambient values (Martinsson), while an additional 10-20% is coming from size cut. There is too much uncertainty to speak about absorption amplification.

This is an estimation from the example shown in figure S6. We agree that there is a lot of uncertainty in this discussion but again, it is important and interesting. The MAC calculated by Martinsson et al. used EC measurements. For example, Pileci et al. reported up to a factor of 2 difference between rBC and EC measurements from several European sites. We report the eBC values using standard Aethalometer settings of MAC. MAC values can certainly be site specific, but also have a seasonal dependence. Zanatta et al. calculated site specific values for a nearby site in southern Sweden that were even lower than the default values we used. We try to discuss potential reasons for the discrepancy but do not have a conclusive answer. We believe that this could be an interesting paper on its own, perhaps using longer datasets and/or lab campaigns.

Pileci, R.E., Modini, R.L., Bertò, M., Yuan, J., Corbin, J.C., Marinoni, A., Henzing, B., Moerman, M.M., Putaud, J.P., Spindler, G. and Wehner, B., 2021. Comparison of co-located refractory black carbon (rBC) and elemental carbon (EC) mass concentration measurements during field campaigns at several European sites. Atmospheric Measurement Techniques, 14(2), pp.1379-1403.

Zanatta, M., Gysel, M., Bukowiecki, N., Müller, T., Weingartner, E., Areskoug, H., Fiebig, M., Yttri, K.E., Mihalopoulos, N., Kouvarakis, G. and Beddows, D., 2016. A European aerosol phenomenology-5: Climatology of black carbon optical properties at 9 regional background sites across Europe. Atmospheric environment, 145, pp.346-364.

L220-223: so why not use the Martinsson MAC? There is no explanation behind the choice of 7.77 m2/g.

See discussion above. The default MAC is what is used by many to report BC.

F2: what is the small window in the plot?

Insert shows the normalized distribution for comparison of the modes (already in caption).

L234: the fact that aerosol diameters are affected by cloud processing is true. I wonder how this is relevant to your study. If this is a general statement, please provide at least some references.

We removed this statement to not add confusion. Now it reads:

L256. The difference in mass GMD is statistically significant and can be explained by different BC particle sources, air masses and coagulation.

F3: legend should simply describe what is shown in the graph. Avoid adding interpretation of results, this belongs to the text. I am not sure what the crosses indicate. I imagine that whiskers are 10-90 percentile…missing

The + is outliers, which is mentioned in the caption. The whiskers show the max and mean of data (as long as not outliers). This is standard for boxplots. We deleted the interpretation so caption now reads:

Figure 4. Box-plot showing the daily pattern of rBC GMD (2 h averages) as measured by the SP2, at the urban and rural sites. Boxes show the median, 25th and 75th percentiles, with values more than 1.5 below or above those considered outliers (+).

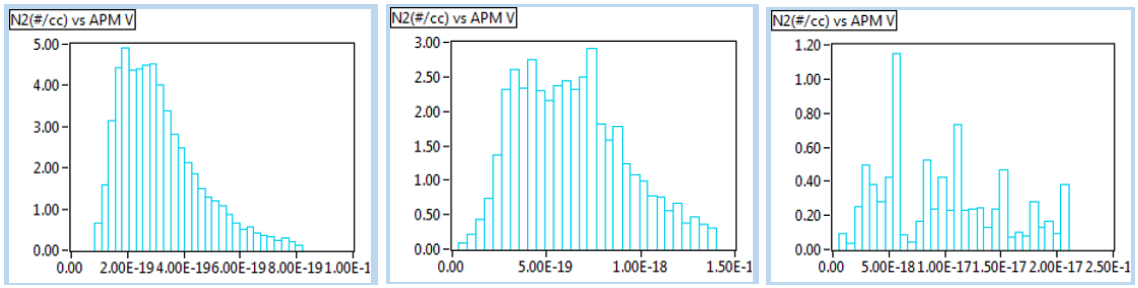
L242: very few people know what is the broadband channel. Since it is not essential information to interpret your result, I would remove it in the result section.

Agree, we removed broadband. Sentence now reads:

L266. The fraction of "thickly coated" BC particles with an incandescence peak height corresponding to a BC core diameter of mass 2.4 fg (~136 nm mass equivalent diameter) was on average 71 ± 8 % (1σ) at the rural site and 38 ± 7 % (1σ) at the urban site, with no big difference during weekends.

L245: these effective density measurements are interesting. You could show the mass distribution for all selected diameters…it would help to understand your text. I might have missed this info, but did you measure effective density at the rural site? Do you see this bimodal distribution?

No, the DMA-APM measurements were only performed in the city. As discussed in section 3.4 we choose to show the 150 nm run since that is where the bimodality was most clear (which is a balance between the fraction of soot particles and total number of particles to get good statistics). Below we show some examples of other sizes (from left to righ: 75 nm, 100 nm, 250 nm). We already included the results of other sizes in the manuscript but don't think showing the other sizes would add to the manuscript main points.



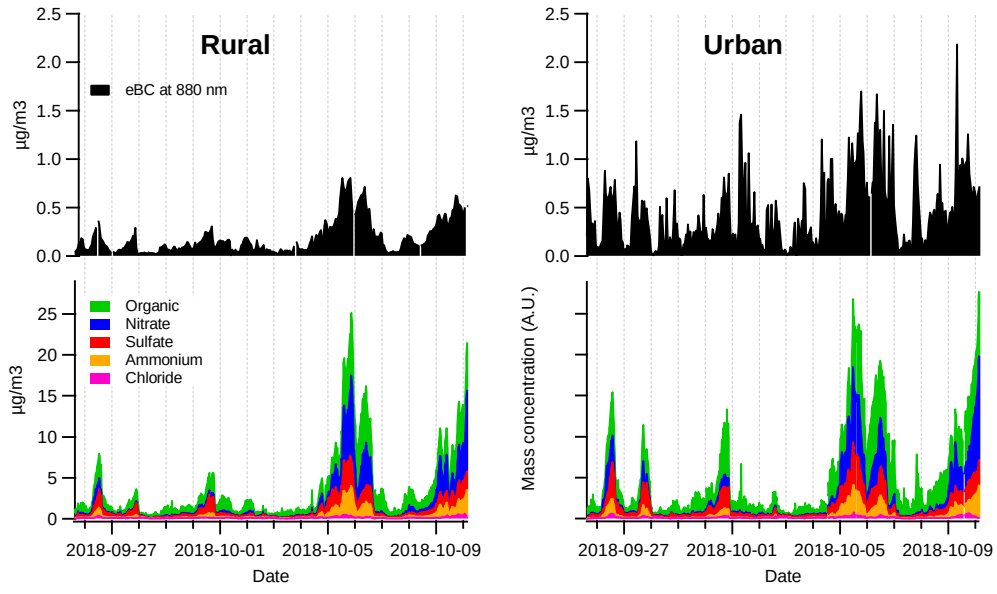
L271: At what altitude the back trajectories are passing over Malmo?

50 m above ground level. Added the following to the manuscript:

L295. From time series comparison with HYSPLIT trajectories, it could be seen that some peaks in eBC, non-refractory PM1, and total particle number concentrations coincide with trajectories passing 50 m above ground level over Malmö/Copenhagen before arrival at the rural site.

F4: what is this arbitrary unit?

Mass concentration. Figure changed:



L275: No actual description is given for the one-year-long dataset. It is very confusing, especially without a dedicated subsection with numbering and title.

We hope that the addition of subsections and graphics above have helped clarify this.

L300-320: did you use the one-year-long dataset for the malmo influence? Not clear to me. Specify the use of the 1year dataset. If not, I am not very persuaded that your 15 days measurements could show or not show any systematic impact of Malmo emission on background aerosol concentration.

We agree that 15 days is short and this is exactly why we also used the one year eBC dataset to further investigate the Malmö/Copenhagen influence. See answers above regarding clarifications.

L331: ok, but why?

See discussion regarding L213 and L217 above.

L339: first-time HR-ToF-AMS is mentioned

We have changed to SP-AMS

L349-352: what are regional traffic sources? You clearly show that the urban environment is impacted by traffic; so, emission reduction policies will benefit the urban population. But it will have a less evident impact on rural locations. I find this final statement a bit confusing, I suggest rephrasing.

We agree that this reads a bit confusing and have rephrased the last paragraph to the following:

L375. The results show that local urban emissions of BC in southern Sweden have a small effect on concentrations and properties in the regional background air. Local abatement strategies aimed towards reducing BC emissions from traffic sources will thus have an effect mostly limited to the urban population. The contribution of long-range transported BC will be even more evident when diesel-driven vehicles are now rapidly being replaced with other forms of propulsion, such as electric motors or hydrogen fuels.

Furthermore we found an erroneous use of the word regional in the results section. L293 in the submitted manuscript was changed from:

L293. It is clear that non refractory-PM1 concentration and composition are driven by regional pollution at both sites, while black carbon is more influenced by local emissions. Regional background sources dominates even the organic aerosol, for which one could expect a larger difference between the sites owing to the high urban traffic density (Glasius et al., 2011).

To:

L318. It is clear that non refractory-PM1 concentration and composition are driven by long-range pollution at both sites, while black carbon is more influenced by local emissions. Long-range sources dominates even the organic aerosol, for which one could expect a larger difference between the sites owing to the high urban traffic density (Glasius et al., 2011).

REVIEWER #2

The article of Ahlberg et al. investigated black carbon concentrations, size distributions, mixing state and sources in two measurement campaigns in Southern Sweden; urban and rural locations. They measured BC by a single particle soot photometer and aethalometer, and complemented the measurements by number size distribution and PM1 chemical composition measurements. In addition to examine the characteristics of BC, the aim of this study was to investigate the contribution of regional sources and long-range transport to the observed BC concentrations.

BC concentrations were larger at the urban site than at the rural site, especially during the traffic rush hour. Also the number of particles having BC core was larger at the urban site and BC particles were smaller in size and had thinner coating. Based on the trajectory analysis, air masses coming from Eastern Europe comprised twice as much BC compared to Western Europe. Nearby Malmö/Copenhagen region impacted rural site BC concentrations only to some extent.

This article presented new results from the field measurements and had enough conclusions for a measurement report. However, the paper is missing a lot of details (for example the measurement sites and periods) and is therefore partly unclear for the reader. This paper merits publication in ACP measurement reports after addressing the comments given below.

General comments

The paper is confusing for the reader as there are so many different measurement periods that are not clearly described in text. For example, the rooftop urban site is only mentioned in one sentence even though the results from the rooftop are shown in supplemental material and discusses in text. Also, the one year measurements for BC are not mentioned in methods section at all. A table or figure listing all sites, instruments and measurement periods would help the reader to get an overall picture of the dataset utilized in the paper.

We hope that the implemented changes discussed above have helped the readers. Specifically the addition of subsections and graphic showing the data used. We did not include the rooftop measurements in this graphic since it was only used to show that the curbside measurements are valid as an indicator of urban BC as discussed in section 3.1 and shown in figure S4.

Specific comments

Title: The title does not cover all the aspects of the text. Based on the title, the study focuses on background air and the effect of regional sources, whereas to me, the object of this paper is much broader. I suggest considering to change the title to reflect better the whole study.

We have tried to change the title into something more suiting. Our new title is "Black carbon properties and concentrations in Southern Sweden urban and rural air – The importance of long-range transport".

Abstract

lines 22-23: …. higher than the background levels… higher than background levels at the urban site or at the rural site? Specify

Clarified that it is compared to rural background (and stitched together with the previous sentence):

L21. Equivalent and refractory BC mass concentrations at the urban site were on average a factor 2.2 and 2.5, with peaks during rush hour up to a factor ~4, higher than the rural background levels.

line 25: fresh plumes of traffic?

Yes, clarified.

L24. The organic components of the fresh traffic plumes were similar in mass spectral signature to "hydrocarbon-like organic aerosol" (HOA), commonly associated with traffic.

line 25: "hydrocarbon-like organic aerosol", why parentheses?

This is a common acronym, especially for AMS-users.

Methods

lines 64 and 70: PM cut-off was different at urban and rural site (PM2.5 vs PM10). How much that impacted the BC results, can you estimate?

Good point. The bias that could have been introduced is that the measured BC concentrations at the urban site was too low. We don't believe a lot of BC mass is present in the larger particles. Especially the locally emitted particles will not have had time to coagulate and grow. Viidanoja et al. (2002) showed that typically more than 90% of BC resided in PM2.5 at an urban site in Finland. Either way, it is highly unlikely that this would affect our conclusions.

Viidanoja, J., Sillanpää, M., Laakia, J., Kerminen, V.M., Hillamo, R., Aarnio, P. and Koskentalo, T., 2002. Organic and black carbon in PM2. 5 and PM10: 1 year of data from an urban site in Helsinki, Finland. Atmospheric Environment, 36(19), pp.3183-3193.

lines 64-65: add details of rooftop measurements

The rooftop measurements are the official Malmö municipality urban background sampling site. We included some details:

L66. The validity of the curbside site measurements as an indicator of the BC concentrations in the city was assessed by comparing simultaneously collected data at a rooftop urban background site (20 m a.g., PM2.5 inlet).

lines 71-72: Why SP2 sampling system is described here only for the rural site?

At the rural site the SP2 sampled through an inlet different from the other instruments. At the urban station all instruments were connected to the described inlet.

line 76: "During the latter part of the urban campaign, chemically resolved particle constituents were measured simultaneously at both sites…" were there simultaneous measurements only for the SP-AMS? Also for AE?

Yes, as can be seen in figure 8. We have clarified that we have used the rural Aethalometer data for the full year in the discussion above.

lines 128-129: It's very difficult to understand why the mass concentrations from the SP-AMS could not be calculated at the urban site as the SMPS number size distributions can be converted to mass size distributions. Were there some issues with the SPMS data as well?

This is also a good point and we tried comparing with both SMPS and TEOM data. For the SMPS, the lower cut-off size meant that the two instruments likely did not see the same mass, and that the mass-size distribution was often cut in the middle of a mode, which introduces a lot of temporal variability. For the TEOM data (not discussed in the manuscript), there was a separate issue with an erroneous baseline. We are still comfortable using the chemical speciation, but not the absolute mass concentrations.

lines 133-139: traffic plumes, did you have any gas monitors (NOx, CO2, CO) that could have been used to indicate traffic plumes as well? How was the diurnal distribution of traffic plumes, were they detected only during rush hours?

There are gas monitors permanently deployed at the urban monitoring site, but we did not look further into this data since plumes were easily discernible using both AMS and SP2. The selected plumes were both from rush hour and night-time.

line 157: APM was run only during five days in spring 2019; how comparable is this data to summer/autumn 2018 data in terms of weather and traffic volume/fleet?

Traffic is rather similar throughout the year, except perhaps during vacation times. Those measurements, to verify the bimodal structure of the aerosol effective density, are just a snapshot that agrees well with the Rissler et al. values.

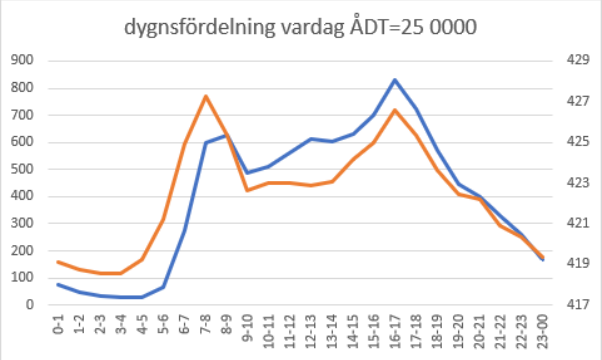
lines 168-170: why the trajectory analysis was carried out only for the rural site, why not both sites?

Our aim was to investigate the origin of BC in the rural background air and specifically if air passing over Malmö had a significant impact. Hence we only used the rural dataset.

Results and discussion

line 184-185: traffic intensity; any figure on that?

Below is a figure showing number of cars (orange, left axis) and CO_2 (blue, right axis) at the site. Data comes from Malmö municipality. We believe this is quite typical behaviour, and not needed in the manuscript.



line 192: "The AAE is similar between the sites with small differences…" To me, 1.13 and 1.24 are not that similar. Could you speculate more the reasons for the difference? Also, AAE is smaller at traffic site than at rural site, is this typical trend?

AAE is actually higher at the traffic site, which may be surprising given the larger contribution of fresh traffic aerosol compared to the rural site. The numbers are significantly different (95% CI), but we don't want to add too much speculation on the reasons, since it concerns short term campaigns that were not simultaneous.

lines 244-257: bimodal distribution, could you discuss more on the sources of two modes?

The two modes comprise of fresh agglomerated soot with a density lower than 1 g/cm^3. The other mode contains more spherical particles (all except soot) which are likely aged. We refer the reader to Rissler et al. (2014) for further reading on the effective density and mixing state of urban aerosol.

lines 270-271: "It was clear that SW winds had more occurrences of high eBC than e.g. NW winds." I somewhat disagree with this sentence. It is clear that the occurrence of SW winds was higher but eBC concentrations are difficult to compare based on Fig. 5 since the occurrence of NW winds is so low. However, Fig 6. shows nicely larger concentration of eBC related to the SE winds.

We agree, it is hard to see in that figure. We removed that sentence.

lines 314-315, correlation of non-refractory-PM1 and thickly coated BC fraction (Fig 8); why 24-hour averaged data with only 15 data points? Why not for example 1-hour averaged data? Was it because of SP2 data?

Yes, the thin/thick coating fraction was evaluated on a 24h basis. In theory it can be done with a higher time-resolution, but this was not implemented in the version of the analysis tool we used.

line 317: non-refractory-PM1 (dominated by secondary material); HOA related to traffic plumes was discussed earlier but there is no data showing that organics were mostly secondary. Could you add some contributions for primary (HOA?) and secondary OA?

In order to quantify our statement that OA at the urban site was dominated by OOA, we used the "m/z tracer method", colloquially known as "poor person's PMF", from Ng et al 2011 stated to reproduce PMF results within 30%. Our result is 78% OOA, 9% HOA for the campaign averaged mass spectrum.

We added the following to the methods:

L150. The m/z tracer method (Ng et al., 2011) was used to estimate the different OA components.

And the following to the results:

L320. The organic aerosol at the urban site was clearly dominated by oxygenated organic aerosol (OOA, 78%) with only a minor contribution of HOA (9%), suggesting that aged long-range sources dominate even the organic aerosol, for which one could expect a larger difference between the sites owing to the high urban traffic density (Glasius et al., 2011).

Ng, N.L., Canagaratna, M.R., Jimenez, J.L., Zhang, Q., Ulbrich, I.M. and Worsnop, D.R., 2011. Real-time methods for estimating organic component mass concentrations from aerosol mass spectrometer data. Environmental science & technology, 45(3), pp.910-916.

Summary and conclusions

lines 341-342 "…but composed of a small fraction of the total aerosol." How much?

9%, see answer right above.

Supplemental material

Fig. S4: Add instruments and time-resolution of data

This is now figure S5. The caption was updated:

Figure S5. Correlation between 1h averages of Aethalometer (eBC) and SP2 (rBC) data.

Technical corrections

line 185: …levels show are similar… correct

Corrected:

L207. The comparison between the urban street-level and urban background eBC levels are similar in time-series but with a lower daily maximum for the roof-top measurements (Fig. S4).

line 339: change HR-ToF-AMS to SP-AMS

Changed.